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(54) **Episulfide group containing alkyl sulfide compounds**

Episulfidgruppen enthaltende Alkylsulfidverbindungen

Composés de sulfide alcoyle contenant des groupes episulfide

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TOATSU CHEM INC)**

Remarks:

The file contains technical information submitted  
after the application was filed and not included in this  
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and a sufficient improvement effect has not been obtained yet.

**[0003]** In the conventional optical materials obtained from the episulfide compounds and the combinations of the polythiol compounds and the isocyanate compounds, the increase in the refractive index is limited, and this increase in the refractive index leads to the deterioration of the Abbe's number. Therefore, there has been a problem that the sufficiently high refractive index and Abbe's number cannot be balanced with each other. Furthermore, the improvement of the above-mentioned optical properties, i.e., the refractive index and the Abbe's number leads to the deterioration of the heat resistance, and therefore there has been a problem that while the sufficiently high refractive index and Abbe's number are balanced with each other, the excellent heat resistance cannot be obtained.

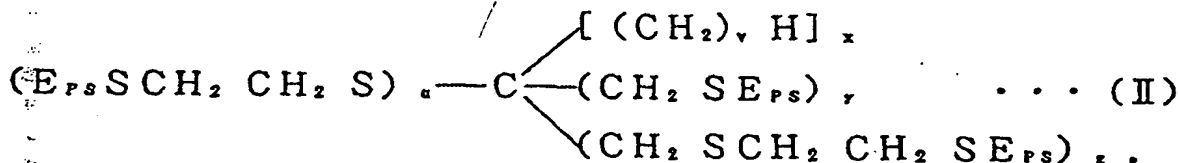
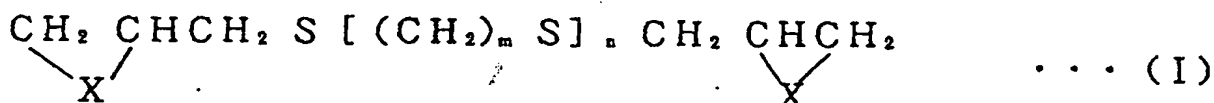
## SUMMARY OF THE INVENTION

**[0004]** An object of the present invention is to provide a novel sulfur-containing compound which can become an optical material having a small thickness and a low chromatic aberration.

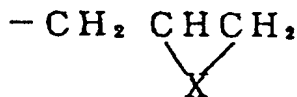
**[0005]** Another object of the present invention is to provide a novel sulfur-containing compound which can become an optical material having a small thickness, a low chromatic aberration and a high heat resistance.

**[0006]** Still another object of the present invention is to provide a novel optical material having such excellent optical properties as mentioned above.

**[0007]** That is to say, the present invention is directed to an alkyl sulfide type episulfide compound represented by the general formula (I) or (II)

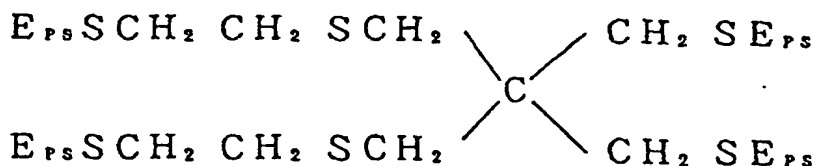


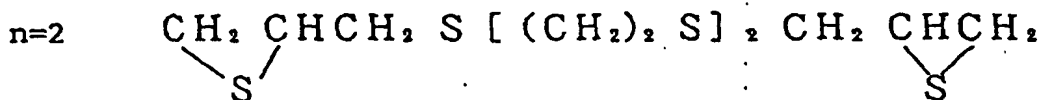
**[0008]** In the general formula (I), m is an integer of 1 to 6; n is an integer of 0 to 4; X is S or O, and at least one X is S. Furthermore, in the general formula (II), x is an integer of 0 to 1; y is an integer of 0 to 4; z is an integer of 0 to 4; u is an integer of 0 to 1; v is an integer of 0 to 3; the relation of  $x+y+z+u=4$  is met; and  $E_{ps}$  is a



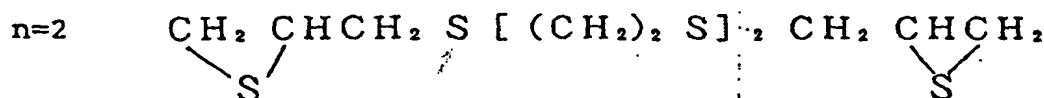
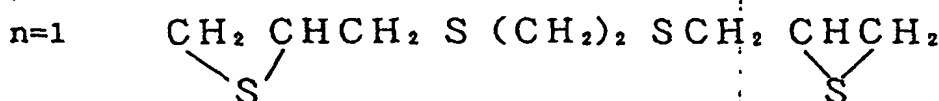
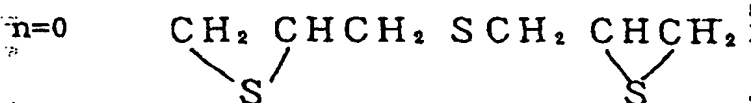
group wherein X is S or O and at least two X are S.

**[0009]** In this general formula (II), for example, in the case that y is plural, y CH<sub>2</sub>SE<sub>ps</sub> bond to carbon atoms, respectively, and similarly in the case that z is plural, z CH<sub>2</sub>SCH<sub>2</sub>CH<sub>2</sub>SE<sub>ps</sub> bond to carbon atoms, respectively. That is to say, for example, in the case of u=0, x=0, y=2 and z=2, the general formula (II) is as follows:





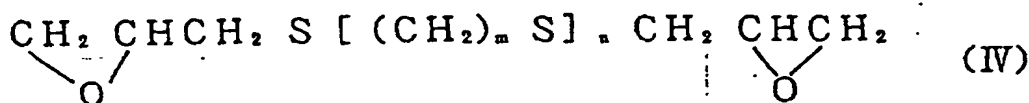
[0012] Among these examples, the compounds represented by the following formulae are most preferable.



[0013] The novel straight-chain alkyl sulfide type epi-sulfide compound of the present invention represented by the general formula (I) can be prepared by various methods, but for example, it can be prepared by reacting a dimercaptan compound having a straight-chain alkyl sulfide structure represented by the general formula (III)



with an epihalohydrin typified by epichlorohydrin in the presence of an alkali to obtain a straight-chain alkyl sulfide type epoxy compound represented by the general formula (IV)



and then reacting this epoxy compound with a sulfurizing agent such as a thiocyanate, thiourea, triphenylphosphine sulfide or 3-methylbenzothiazole-2-thione, preferably the thiocyanate or thiourea. In preparing the epoxy compound represented by the above-mentioned general formula (IV), epichlorohydrin is preferable as the epihalohydrin compound. Furthermore, the epihalohydrin compound is stoichiometrically used in an amount of 2 mol per mol of the dimercaptan compound having the general formula (III), but in view of the purity, the reaction rate, the economy and the like of the product, the epihalohydrin compound may be used in a more amount or in a less amount than described above. The epihalohydrin compound is preferably used for the reaction in an amount of 2 to 10 mol, more preferably 2 to 5 mol per mol of the dimercaptan compound. The reaction may be carried out in the absence or presence of a solvent, but when the solvent is used, it is desirable to use the solvent in which any one of the epihalohydrin, the dimercaptan compound of the general formula (III) or a metallic salt of the dimercaptan compound is soluble. Typical examples of such a solvent include water, alcohols, ethers, aromatic hydrocarbons, halogenated hydrocarbons and mixtures thereof. The reaction easily proceeds in the presence of a base. Examples of the base include pyridine, tertiary amines such as triethylamine and diaza-bicyclo-undecene, and hydroxides of alkali metals and alkaline earth metals. Above all, the hydroxides of the alkali metals and the alkaline earth metals are preferable, and sodium hydroxide and

[0017] Above all, the cases (a) of  $x=0$  and the cases (b) to (d) of  $x=1$  and  $v=0$  to 2 are preferable, and in the case (a) of  $x=0$  (a), the following compounds of  $u=0$  and  $y+z=4$  are more preferable:

( $x=0, y=4, z=0, u=0$ ), ( $x=0, y=3, z=1, u=0$ ), ( $x=0, y=2, z=2, u=0$ ), ( $x=0, y=1, z=3, u=0$ ), ( $x=0, y=0, z=4, u=0$ ).

[0018] In the case (b) of  $x=1$  and  $v=0$ , the following compounds of  $u=1$  and  $y+z=2$  are preferable:

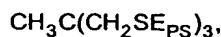
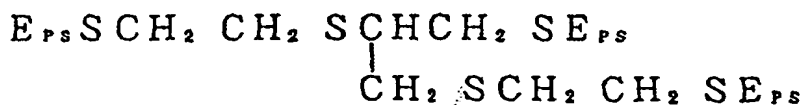
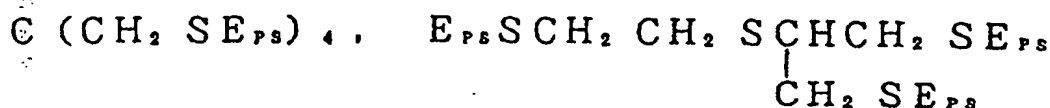
( $x=1, y=2, z=0, u=1$ ), ( $x=1, y=1, z=1, u=1$ ), ( $x=1, y=0, z=2, u=1$ ).

[0019] In the cases (c) and (d) of  $x=1$  and  $v=1$  to 2, the following compounds of  $u=0$  and  $y+z=3$  are preferable:

( $x=1, y=3, z=0, u=0$ ), ( $x=1, y=2, z=1, u=0$ ), ( $x=1, y=1, z=2, u=0$ ), and ( $x=1, y=0, z=3, u=0$ ).

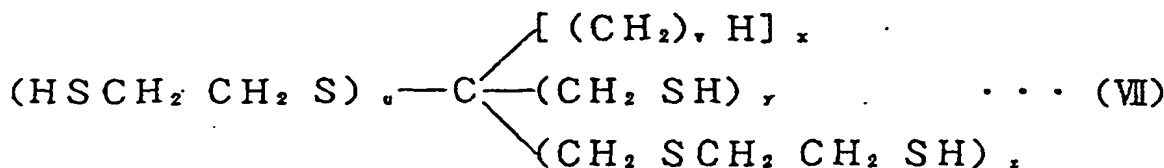
[0020] Most preferable are ( $x=0, y=4, z=0, u=0$ ) in the case (a) of  $x=0$ ; ( $x=1, y=2, z=0, u=1$ ) and ( $x=1, y=1, z=1, u=1$ ) in the case (b) of  $x=1$  and  $v=0$ ; and ( $x=1, y=3, z=0, u=0$ ) in the cases (c) and (d) of  $x=1$  and  $v=1$  to 2.

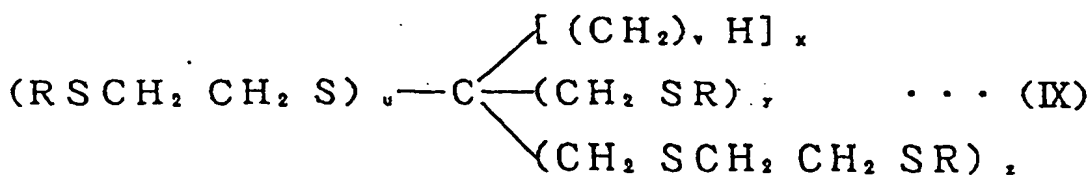
[0021] The most preferable examples of the compound represented by the general formula (II) will be enumerated by structural formulae in the above-mentioned order as follows.



[0022] In the above-mentioned general formula (II), X is S or O, as in the above-mentioned general formula (I), but at least two X are S, preferably all X are S. The preferable examples, the more preferable examples and the most preferable examples of the novel branched alkyl sulfide type episulfide compound represented by the general formula (II) of the present invention have hereinbefore been shown, but the grounds of such a ranking are as follows. If the molecular weight of a branch portion constituting a branch of the episulfide compound is too large, the heat resistance of the optical material which can be obtained by polymerizing/curing the compound cannot sufficiently be exerted, and if a sulfur content is low, the sufficiently high refractive index cannot be obtained (this tendency is particularly noticeable, when the compound is polymerized/cured together with another compound). Furthermore, if the number of S in X is less than two, the sulfur content is low, so that the high refractive index cannot be achieved. In addition, the reactivity of the compound deteriorates, so that high-temperature conditions are necessary for the polymerization, with the result that the optical material is inconveniently colored. Moreover, another ground is that if a similar effect is obtained, the structure not having many branches is preferable for the sake of avoiding the complexity of the preparation process.

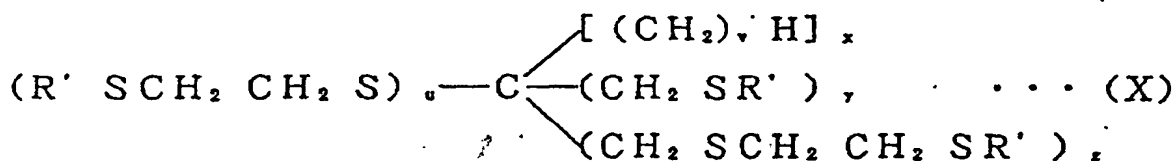
[0023] The novel branched alkyl sulfide type episulfide compound represented by the general formula (II) of the present invention can be prepared by various methods, but for example, a trimercaptan compound or a tetramercaptan compound having a branched alkyl sulfide structure represented by the following general formula (VII) is reacted with an epihalohydrin typified by epichlorohydrin in the presence of an alkali to obtain a branched alkyl sulfide type epoxy compound represented by the following general formula (VIII):





wherein x is an integer of 0 or 1; y is an integer of 0 to 4; z is an integer of 0 to 4; u is an integer of 0 or 1; and v is an integer of 0 to 3; the relation of  $x+y+z+u=4$  is met; and R is a  $CH_2=CHCH_2-$  group.

[0025] The unsaturated compound of the general formula (IX) can be prepared by, for example, condensing the branched dimercaptan compound of the general formula (VII) and an allyl halide compound such as allyl chloride or allyl bromide in the presence of a base. Furthermore, another effective process is also present which comprises the dehalogenation-hydrogenation reaction of a trihalotrimercaptan compound represented by the following general formula (X) or a tetrahalotetramercaptan compound:



wherein x is an integer of 0 or 1; y is an integer of 0 to 4; z is an integer of 0 to 4; u is an integer of 0 or 1; and v is an integer of 0 to 3; the relation of  $x+y+z+u=4$  is met; R' is a  $X^1CH_2CHSHCH_2-$  group, and  $X^1$  is a chlorine atom or a bromine atom.

[0026] It is known that the halomercaptan compound of the general formula (X) can easily be synthesized from the unsaturated compound of the general formula (IX) and sulfur chloride or the like [e.g., F. Lautenschlaeger et al., J. Org. Chem., Vol. 34, p. 396 (1969)].

[0027] One or more of the novel alkyl sulfide type episulfide compounds of the present invention [the straight-chain type of the general formula (I) and the branched type of the general formula (II)] can be polymerized/cured in the presence of a curing catalyst to prepare an optical material. In this polymerization, any of the known curing catalysts for epoxy resins can be used. Typical examples of the curing catalysts include

(1) amine compounds typified by primary amines such as ethylamine, n-propylamine, sec-propylamine, n-butylamine, sec-butylamine, iso-butylamine, tert-butylamine, pentylamine, hexylamine, heptylamine, octylamine, decylamine, laurylamine, myristylamine, 1,2-dimethylhexylamine, 3-pentylamine, 2-ethylhexylamine, allylamine, aminoethanol, 1-aminopropanol, 2-aminopropanol, aminobutanol, aminopentanol, aminohexanol, 3-ethoxypropylamine, 3-propoxypropylamine, 3-isopropoxypropylamine, 3-butoxypropylamine, 3-isobutoxypropylamine, 3-(2-ethylhexyloxy)propylamine, aminocyclopentane, aminocyclohexane, aminonorbornene, aminomethylcyclohexane, aminobenzene, benzylamine, phenethylamine,  $\alpha$ -phenylethylamine, naphthylamine and furfurylamine; primary polyamines such as ethylenediamine, 1,2-diaminopropane, 1,3-diaminopropane, 1,2-diaminobutane, 1,3-diaminobutane, 1,4-diaminobutane, 1,5-diaminopentane, 1,6-diaminohexane, 1,7-diaminoheptane, 1,8-diaminooctane, dimethylaminopropylamine, diethylaminopropylamine, bis-(3-aminopropyl) ether, 1,2-bis-(3-aminopropoxy)ethane, 1,3-bis-(3-aminopropoxy)-2,2'-dimethylpropane, amonoethylethanolamine, 1,2-bisaminocyclohexane, 1,3-bis-aminocyclohexane, 1,4-bisaminocyclohexane, 1,3-bisaminomethylcyclohexane, 1,4-bisaminomethylcyclohexane, 1,3-bisaminoethylcyclohexane, 1,4-bisaminoethylcyclohexane, 1,3-bisaminopropylcyclohexane, 1,4-bisaminopropylcyclohexane, hydrogenated 4,4'-diaminodiphenylmethane, 2-aminopiperidine, 4-aminopiperidine, 2-aminomethylpiperidine, 4-aminomethylpiperidine, 2-aminoethylpiperidine, 4-aminoethylpiperidine, N-aminoethylpiperidine, N-aminopropylpiperidine, N-aminoethylmorpholine, N-aminopropylmorpholine, isophoronediamine, methanediamine, 1,4-bisaminopropylpiperazine, o-phenylenediamine, m-phenylenediamine, p-phenylenediamine, 2,4-tolylenediamine, 2,6-tolylenediamine, 2,4-toluenediamine, m-aminobenzylamine, 4-chloro-o-phenylenediamine, tetrachloro-p-xylylenediamine, 4-methoxy-6-methyl-m-phenylenediamine, m-xylylenediamine, p-xylylenediamine, 1,5-naphthalenediamine, 2,6-naphthalenediamine, benzidine, 4,4'-bis(o-toluidine), dianisidine, 4,4'-diaminodiphenylmethane, 2,2-(4,4'-diaminodiphenyl)-propane, 4,4'-diamino

the cured material from a mold. Examples of the internal release agent referred to herein include fluorine-containing nonionic surface active agents, silicon-containing nonionic surface active agents, alkyl quaternary ammonium salts, phosphoric acid esters, acidic phosphoric acid esters, alkali metal salts of the acidic phosphoric acid esters, metal salts of higher fatty acids, higher fatty acid esters, paraffins, waxes, higher aliphatic amides, higher aliphatic alcohols, polysiloxanes and adducts of aliphatic amines with ethylene oxides.

**[0031]** In the case that the novel alkyl sulfide type episulfide compound of the present invention as a starting material is polymerized/cured, if necessary, by the use of the above-mentioned curing catalyst and a compound such as glycidyl methacrylate which can react with an episulfide group having an unsaturated group in order to obtain an optical material, these materials are mixed with additives such as a radical polymerization initiator, a radically polymerizable monomer, a release agent, an antioxidant and an ultraviolet light absorber, and the mixture is then polymerized/cured by the following procedure to obtain an optical material such as a lens. That is to say, the mixed material is poured into a glass mold or a metal mold, and a polymerizing/curing reaction is then allowed to proceed by heating. Afterward, the thus cured material is released from the mold. A curing time is in the range of 0.1 to 100 hours, usually 1 to 48 hours, and a curing temperature is in the range of -10 to 160°C, usually -10 to 140°C. Alternatively, after the completion of the curing, the material may be subjected to an annealing treatment at a temperature of 50 to 150°C for a period of 10 minutes to 5 hours, and this annealing treatment is preferable to remove strain from the optical material of the present invention. Furthermore, if necessary, a surface treatment suitable for the formation of a hard coat, the prevention of reflection, the impartment of fog resistance or the like can selectively be carried out.

**[0032]** Next, the present invention will be described in detail with reference to examples, but the scope of the present invention should not be limited to these examples. Incidentally, the physical properties of obtained polymers were measured by the following procedures.

**[0033]** Refractive index ( $N_D$ ) and Abbe's number ( $v_D$ ): They were measured at 25°C by the use of an Abbe's refractometer.

**[0034]** Specific gravity: It was measured at 25°C by the use of an electron gravimeter, and then calibrated in a usual manner.

**[0035]** Heat resistance: A product having a Vicat softening point of 120°C or more was represented by O, a product having a softening point of less than 120°C and 80°C or more was represented by Δ, and a product having a softening point of less than 80°C was represented by ×.

**[0036]** Strength: In accordance with a three-point bending test using an autograph, a product having a bend of 10 mm or more was represented by O, a product having a bend of less than 10 mm and 5 mm or more was represented by Δ, and a product having a bend of less than 5 mm was represented by ×.

#### Example 1 (in the general formula (I), $n=1$ and $m=2$ )

**[0037]** In a flask equipped with a stirrer, a dropping funnel, a thermometer and a nitrogen introducing tube were placed 94.2 g (1.0 mol) of 1,2-dimercaptoethane, an aqueous solution obtained by dissolving 0.4 g (10 mmol) of sodium hydroxide in 4 ml of water and 100 ml of methanol, and 2.0 mol (185.0 g) of epichlorohydrin was then added dropwise over 1 hour. During the dropping, a reaction temperature was maintained at 0 to 10°C. After the completion of the dropping, the reaction was further continued for 1 hour. Afterward, 240 g (6.0 mol) of caustic soda dissolved in 360 ml of water was added dropwise over 1 hour. During the dropping, a reaction temperature was maintained at 0 to 10°C. After the completion of the dropping, the reaction was further continued for 3 hours. After extraction with toluene and washing with water, the used solvent was distilled off to obtain 202.0 g (99% of a theoretical amount) of 1,2-bis(glycidylthio)ethane in the state of a colorless transparent liquid.

**[0038]** Next, in a flask equipped with a stirrer, a thermometer and a nitrogen introducing tube were placed 79.9 g (0.3 mol) of the thus obtained 1,2-bis(glycidylthio)ethane, 95.3 g (1.25 mol) of thiourea, 2.96 g (0.03 mol) of acetic anhydride and 250 ml of toluene as well as 250 ml of methanol as solvents, and reaction was then carried out at 30°C for 9 hours. After the reaction, the reaction solution was extracted with toluene, and the resulting extract was washed with a 1% aqueous sulfuric acid solution and then water. Afterward, the excessive solvents were distilled off to obtain 82.0 g (93% of a theoretical amount) of 1,2-bis (β-epithiopropylthio) ethane (in the general formula (I),  $m=2$  and  $n=1$ ) in the state of a white solid.

Elemental analysis:	Found	Calcd.
C	40.19%	40.30%
H	6.05%	5.95%
S	53.79%	53.64%

Mass spectrum (EI):

$M^+$  238 (theoretical molecular weight = 238)

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Mass spectrum (EI):  $M^+$  298 (theoretical molecular weight = 298)  
 Infrared absorption spectrum:  $620\text{ cm}^{-1}$  (stretching vibration of an episulfide ring)  
 $^1\text{H-NMR}$ :  
     2.2 ppm (hydrogen atoms bonded to carbon atoms at 1- and 13-positions)  
     2.6 ppm (hydrogen atoms bonded to carbon atoms at 1- and 13-positions)  
     2.7 ppm (hydrogen atoms bonded to carbon atoms at 2- and 12-positions)  
     2.8-2.9 ppm (hydrogen atoms bonded to carbon atoms at 5-, 6-, 8- and 9-positions)  
     3.0 ppm (hydrogen atoms bonded to carbon atoms at 3- and 11-positions)  
     3.1 ppm (hydrogen atoms bonded to carbon atoms at 3- and 11-positions)  
 $^{13}\text{C-NMR}$ :  
     25.7 ppm (carbon atoms at 1- and 13-positions)  
     32.4 ppm (carbon atoms at 2- and 12-positions)  
     32.5 ppm (carbon atoms at 6- and 8-positions)  
     34.0 ppm (carbon atoms at 5- and 9-positions)  
     38.4 ppm (carbon atoms at 3- and 11-positions)

**[0043]** After polymerization/curing, the refractive index, the Abbe's number and the specific gravity of an obtained optical material were measured, and the results are shown in Table 1.

### Example 4 (in the general formula (I), $n=0$ )

**[0044]** 1.0 mol (34.1 g) of hydrogen sulfide and 1.0 mol (92.5 g) of epichlorohydrin were placed in a completely sealed reaction device, and the solution was cooled to  $10^\circ\text{C}$ . Next, an aqueous solution obtained by dissolving 5 mmol (0.2 g) of an aqueous sodium hydroxide solution in 4 ml of water was added to the cooled solution, followed by stirring at this temperature for 1 hour. Afterward, the solution was stirred for 2 hours, while the solution temperature was maintained at about  $40$  to  $45^\circ\text{C}$ . The solution was cooled to  $10^\circ\text{C}$  again, and an aqueous solution obtained by dissolving 1.0 mol (92.5 g) of epichlorohydrin and 5 mmol (0.2 g) of sodium hydroxide in 4 ml of water was further added to the cooled solution, followed by stirring at  $10^\circ\text{C}$  for 1 hour and at  $40$  to  $45^\circ\text{C}$  for 2 hours. After the solution temperature had been returned to room temperature, an aqueous solution obtained by dissolving 80.0 g (2 mol) of sodium hydroxide in 80 ml of water was added dropwise thereto, while the solution temperature was maintained at about  $40$  to  $45^\circ\text{C}$ , followed by stirring for 3 hours, while the solution temperature was maintained at about  $40$  to  $45^\circ\text{C}$ . After the completion of the reaction, the same treatment as in Example 1 was made to obtain 92.2 g (theoretical amount = 63%) of a product. Next, this product was reacted with thiourea to obtain bis( $\beta$ -epithiopropyl) sulfide (in the general formula (I),  $n=0$ ) in a yield of 80%.

Elemental analysis:	Found	Calcd.
C	40.25%	40.41%
H	5.81%	5.65%
S	53.77%	53.94%

Mass spectrum (EI):  $M^+$  178 (theoretical molecular weight = 178)  
 Infrared absorption spectrum:  $620\text{ cm}^{-1}$  (stretching vibration of an episulfide ring)  
 $^1\text{H-NMR}$ :  
     2.3 ppm (hydrogen atoms bonded to carbon atoms at 1- and 7-positions)  
     2.6 ppm (hydrogen atoms bonded to carbon atoms at 1- and 7-positions)  
     2.7 ppm (hydrogen atoms bonded to carbon atoms at 2- and 6-positions)  
     3.0-3.1 ppm (hydrogen atoms bonded to carbon atoms at 3- and 5-positions)  
 $^{13}\text{C-NMR}$ :  
     25.6 ppm (carbon atoms at 1- and 7-positions)  
     33.8 ppm (carbon atoms at 2- and 6-positions)  
     38.6 ppm (carbon atoms at 3- and 5-positions)

**[0045]** After polymerization/curing, the refractive index, the Abbe's number and the specific gravity of an obtained optical material were measured, and the results are shown in Table 1.

### Example 5

**[0046]** The same procedure as in Example 1 was repeated except that 1,2-dimercaptoethane was replaced with 1,6-dimercaptohexane, thereby obtaining 1,6-bis( $\beta$ -epithiopropylthio)hexane in a total yield of 72%.

Table 1

	Episulfide Compound	N <sub>D</sub>	$\nu_D$	Specific Gravity	Heat Resistance	Strength
Example 1	1,2-bis( $\beta$ -epithio- propylthio)ethane	1.70	36	1.38	O	O
Example 2	1,4-bis( $\beta$ -epithio- propylthio)butane	1.68	37	1.32	O	O
Example 3	Bis( $\beta$ -epithiopropyl- thioethyl) sulfide	1.70	36	1.35	O	O
Example 4	Bis( $\beta$ -epithio- propyl) sulfide	1.71	35	1.40	O	$\Delta$
Example 5	1,6-bis( $\beta$ -epithio- propylthio)hexane	1.67	38	1.30	$\Delta$	O
Comp. Ex. 1	1,2-bis( $\beta$ -epithio- propyloxy)ethane	1.60	42	1.30	$\Delta$	$\Delta$
Comp. Ex. 2	1,4-bis( $\beta$ -epithio- propyloxy)butane	1.59	43	1.26	$\Delta$	$\Delta$
Comp. Ex. 3	Bis( $\beta$ -epithiopropo- pyloxyethyl) ether	1.58	43	1.27	X	O
Comp. Ex. 4	In general formula (I) of n=0 and m=2, ratio of S in X was 30% of total of S and O on the average.	1.63	41	1.33	X	O



**Example 8** (in the general formula (II),  $x=0$ ,  $y=4$ ,  $z=0$  and  $u=0$ )

**[0056]** The same procedure as in Example 6 was repeated except that 2-(2-mercaptoethylthio)-1,3-dimercaptopropane was replaced with tetrakis(mercaptomethyl)methane, thereby obtaining tetrakis( $\beta$ -epithiopropylthiomethyl)methane in a total yield of 78%.

Elemental analysis:	Found	Calcd.
C	41.59%	41.76%
H	5.91%	5.77%
S	52.30%	52.47%

Mass spectrum (EI):

$M^+$  488 (theoretical molecular weight = 488)

Infrared absorption spectrum:

620  $\text{cm}^{-1}$  (stretching vibration of an episulfide ring)

$^1\text{H-NMR}$ :

2.2 ppm (hydrogen atoms bonded to carbon atoms at 1-, 11-, 11'- and 11''-positions)

2.6 ppm (hydrogen atoms bonded to carbon atoms at 1-, 5-, 7-, 11-, 7'-, 11'-, 7''- and 11''-positions)

2.7 ppm (hydrogen atoms bonded to carbon atoms at 2-, 10-, 10'- and 10''-positions)

3.0 ppm (hydrogen atoms bonded to carbon atoms at 3-, 9-, 9'- and 9''-positions)

3.1 ppm (hydrogen atoms bonded to carbon atoms at 3-, 9-, 9'- and 9''-positions)

$^{13}\text{C-NMR}$ :

25.7 ppm (carbon atoms at 1-, 11-, 11'- and 11''-positions)

32.5 ppm (carbon atoms at 2-, 10-, 10'- and 10''-positions)

37.3 ppm (carbon atoms at 5-, 7-, 7'- and 7''-positions)

38.5 ppm (a carbon atom at 6-position)

39.1 ppm (carbon atoms at 3-, 9-, 9'- and 9''-positions)

**[0057]** After polymerization/curing, the refractive index, the Abbe's number and the specific gravity of the obtained optical material were measured, and the results are shown in Table 2.

**Example 9** (in the general formula (II),  $v=2$  ( $x=1$ ,  $y=3$ ,  $z=0$  and  $u=0$ ))

**[0058]** The same procedure as in Example 6 was repeated except that 2-(2-mercaptoethylthio)-1,3-dimercaptopropane was replaced with 1,1,1-tris(mercaptomethyl)propane, thereby obtaining 1,1,1-tris( $\beta$ -epithiopropylthiomethyl)propane (in the general formula (II),  $m=2$  and  $v=2$ ) in a total yield of 75%.

Elemental analysis:	Found	Calcd.
C	45.00%	45.18%
H	6.67%	6.57%
S	48.09%	48.25%

Mass spectrum (EI):

$M^+$  398 (theoretical molecular weight = 398)

Infrared absorption spectrum:

620  $\text{cm}^{-1}$  (stretching vibration of an episulfide ring)

$^1\text{H-NMR}$ :

1.0 ppm (hydrogen atoms bonded to a carbon atom at 8''-position)

1.5 ppm (hydrogen atoms bonded to a carbon atom at 7''-position)

2.2 ppm (hydrogen atoms bonded to carbon atoms at 1-, 11- and 11'-positions)

2.6-2.8 ppm (hydrogen atoms bonded to carbon atoms at 1-, 2-, 5-, 7-, 10-, 11-, 7'-, 10'- and 11'-positions)

3.0 ppm (hydrogen atoms bonded to carbon atoms at 3-, 9- and 9'-positions)

3.1 ppm (hydrogen atoms bonded to carbon atoms at 3-, 9- and 9'-positions)

$^{13}\text{C-NMR}$ :

11.4 ppm (a carbon atom at 8''-position)

25.7 ppm (carbon atoms at 1-, 11- and 11'-positions)

28.5 ppm (a carbon atom at 7''-position) 32.4 ppm (carbon atoms at 2-, 10- and 10'-positions)

37.9 ppm (carbon atoms at 5-, 7- and 7'-positions)

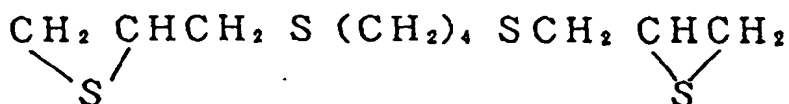
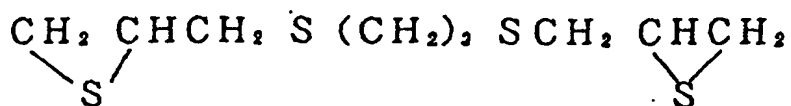
38.8 ppm (a carbon atom at 6-position)

39.1 ppm (carbon atoms at 3-, 9- and 9'-positions)

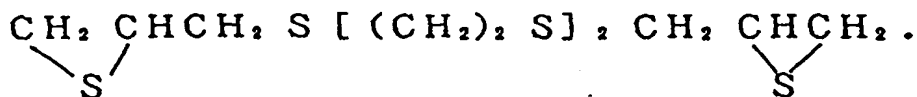
Table 2

Episulfide Compound	$N_D$	$\nu_D$	Specific Gravity	Heat Resistance	Strength
Example 6 2-(2- $\beta$ -epithio-propylthioethyl-thio)-1,3-bis-( $\beta$ -epithiopropylthio)propane	1.69	36	1.38	O	O
Example 7 1,2-bis[(2- $\beta$ -epithio-propylthioethyl-thio)-3-( $\beta$ -epithiopropylthio)propane	1.70	36	1.38	O	O
Example 8 Tetrakis( $\beta$ -epithio-propylthioethyl)methane	1.69	36	1.37	O	O
Example 9 1,1,1-tris( $\beta$ -epithiopropylthio-methyl)propane	1.68	37	1.35	O	O
Comp. Ex. 5 0.2 mol of 2-(2-mercaptoethylthio)-1,3-dimercaptopropane and 0.3 mol of meta-xylylene diisocyanate	1.66	32	1.37	X	O
Comp. Ex. 6 In general formula (II) of $v=0$ , $x=1$ , $y=2$ , $z=0$ and $u=1$ , ratio of S in X was 30% of total of S and 0 on the average.	1.62	38	1.29	$\Delta$	$\Delta$

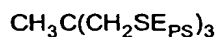
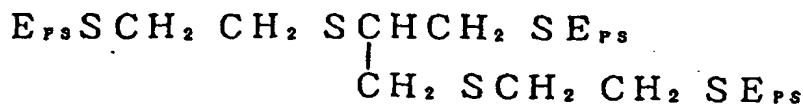
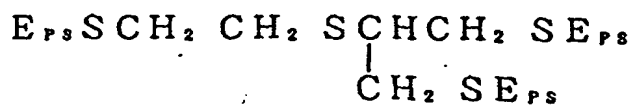
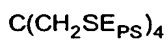
[0062] A novel alkyl sulfide type episulfide compound of the present invention enables the provision of a resinous optical material having a sufficiently high refractive index and a good balance between the refractive index and an Abbe's number which can scarcely be attained by using conventional compounds as materials. In addition, the present invention can provide a resinous optical material having the good balance between the refractive index and the Abbe's number as well as a high heat resistance. In particular, a branched alkyl sulfide type episulfide compound represented



or



**3. The alkyl sulfide type episulfide compound according to Claim 1 which is represented by**



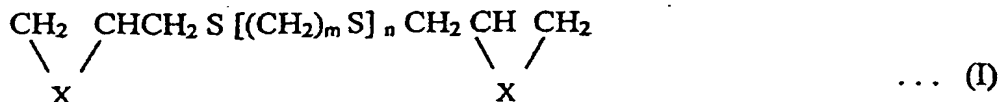
**or**



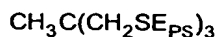
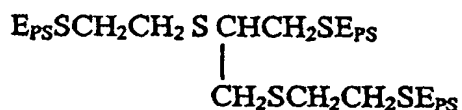
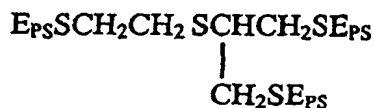
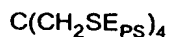
4. An optical material which is obtainable by polymerizing/curing an alkyl sulfide type episulfide compound described in any one of Claims 1 to 3.

## Patentansprüche

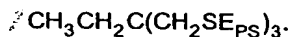
1. Eine Episulfidverbindung vom Alkylsulfidtyp, dargestellt durch die allgemeine Formel (I) oder (II),



3. Die Episulfidverbindung vom Alkylsulfidtyp gemäß Anspruch 1, dargestellt durch



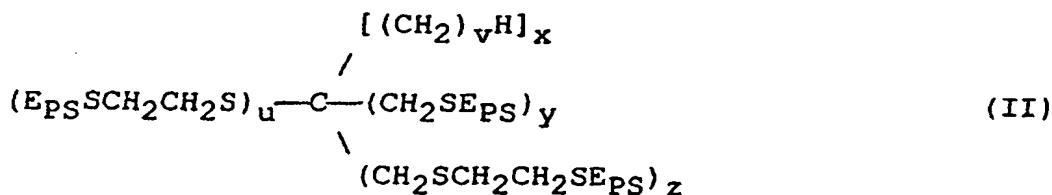
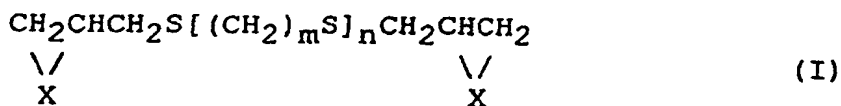
oder



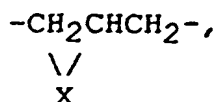
4. Ein optisches Material, das durch Polymerisieren/Härten von einer in mindestens einem der Ansprüche 1 bis 3 beschriebenen Episulfidverbindung vom Alkylsulfidtyp erhältlich ist.

#### Revendications

1. Composé episulfure de type sulfure d'alkyle représenté par la formule générale (I) ou (II):



dans la formule générale (I), m est un nombre entier de 1 à 6 ; n est un nombre entier de 0 à 4 ; X représente un atome S ou O, et au moins un X représente un atome S ; et dans la formule générale (II), x est un nombre entier de 0 à 1 ; y est un nombre entier de 0 à 4 ; z est un nombre entier de 0 à 4 ; u est un nombre entier de 0 à 1 ; v est un nombre entier de 0 à 3 ; la relation  $x+y+z+u=4$  est satisfaite ; et  $E_{ps}$  est un groupe



ou



5

4. Matière optique que l'on peut obtenir en polymérisant/durcissant un composé episulfure de type sulfure d'alkyle décrit selon l'une quelconque des revendications 1 à 3.

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